

0138A-EC-US

Moreover, there is no way that the cumbersome, multi-step process for producing supported catalyst in Kemp et al. could be performed within a continuous gas stream as is the case with the present invention. This multi-step, non-gas stream process is described in the paragraph Col. 1 line 56-Col. 2, line 3:

"In further accord with the invention, a fuel cell electrode catalyst comprising crystals of a noble metal fuel cell catalyst specie deposited on particles of an inert carrier or support powder is prepared by (a) mixing a slurry of inert carrier powder and a solution of a salt of the fuel cell catalyst specie, (b) concentrating and drying the solution, (c) decomposing the platinum salt to form a partial product powder of less than a desired concentration of small catalyst crystals adhered to the inert carrier particles, (d) mixing a slurry of said partial product powder and a solution of a salt of said fuel cell catalyst, (e) repeating step (b), (f) decomposing the salt to form a powder of a higher concentration of catalyst, and (g) repeating steps (d)-(f) as necessary to form a final product powder of a desired concentration of catalyst on carrier particles."

It should be noted that none of the steps of forming the supported catalyst in Kemp et al. are performed in a gas stream. Furthermore, the entire Kemp et al. process takes many hours to complete, particularly because a single processing of the support material with the catalyst precursor does not provide sufficient catalyst loading. This requires multiple repeats of the process. Four repeats of the process are required in the process described in the Example from Col. 2, line 43-Col. 3., line 23. By applicants' calculation, this Example took at least 22 hours to perform and thereby produce the supported catalyst. In contrast, in applicants' process, formation of the supported catalyst takes place in a matter of seconds, and the whole process, from initial introduction of support particulates to deposition of the catalytic layer, take place in under a minute, generally far less than a minute.

It is inherent in Applicants' process that all of the catalyst will deposit on the surface of the support and thereby be available for catalytic activity. On the other hand, in Kemp et al., the process must be repeated several times to achieve the desired catalytic loading. In each repeat, the support and the precursor of the catalyst are mixed, dried into a cake, and the cake then ground. It must be presumed that in each step, a certain amount of the catalyst becomes buried within the particles that are produced, making that portion of the catalyst unavailable for catalytic activity. As catalytic metals tend to be expensive, the processes of the present invention inherently produces a supported catalyst with higher catalytic activity relative to amount of catalyst used.

While the Kemp et al. example added 5 wt% catalyst relative to support particles per cycle, i.e., two cycles to achieve 10%, four to achieve their desired 20%, Applicants' process inherently requires that their desired catalyst loading on support materials occur on a single pass, as once the support/catalyst particles are admixed with the polymer, no further catalyst loading can occur. While Applicants can easily achieve 20% catalyst relative to support loading, even up to 100% (equal weight of catalyst and support), more typically Applicants aim for somewhat greater than 10% catalyst relative to support. Because Applicants' process produces supported catalyst with very small (high surface

0138A-EC-US

area) catalyst particles, all of which are on the support surface, Applicants' target catalyst is typically somewhat lower than the 20% loading sought and achieved through four cycles in the Kemp et al. example. A lower target loading, of course, is more efficient use of expensive metals, particularly platinum.

Applicant's process forms the supported catalytic material, mixes it with polymer, and deposits the mixture to form a catalytic layer, all in a single, continuous gas stream process. For this reason, Claim 33, directed to a method, and Claim 40, directed to apparatus, are believed to be allowable. With regard to *in re Tatincloux 108 USPQ 125*, those claims which involved merely performing simultaneous operations that had previously been performed in sequence were not deemed patentable, but those claims that added to the combination of the previous processes were deemed patentable. Here, the novel and non-obvious efficient process is not just simultaneous performance of prior art processes, but a continuous gas stream process that achieves or exceeds the results of the combination of the prior art processes, neither of which alone is performed entirely in a continuous gas stream.

Neither Kemp nor Singer teach a flowing gas stream in which supported catalyst is produced in a first region of higher temperature and then mixed with polymeric material in a second lower temperature region as per dependent Claim 34, and this claim is believed to be further allowable, as are Claims 35-37 depending therefrom.

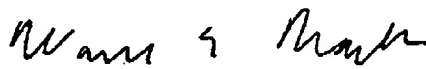
The quenching means of Claims 38 (and 39) are irrelevant to Kemp and Singer and are likewise believed further allowable.

Regarding Claims 34-37, Hunt et al. teach deposition of materials as layers on substrates, but not material on gas-carried particulates. Thus, these claims distinguish over Hunt et al.

For similar reasons claims depending from apparatus Claim 40 are believed to be further allowable.

It is believed all of the claims are currently in condition for allowance; favorable action is courteously requested.

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Wayne E. Nacker, Reg. No. 29,571
Customer No. 24948

0138A-EC-US

APPENDIX A

Claims 1-32 (cancelled)

33. A method for depositing a catalytic layer on a surface of a substrate, the method comprising

providing a continuous gas stream that flows from upstream to downstream to impinge on said substrate,

introducing particulates of support material into said gas stream and, at a first upstream region of said gas stream, depositing catalytic material on said support material particulates to produce particulates of supported catalytic material,

in a second region of said gas stream downstream of said first region, providing a spray comprising polymeric material so as to mix said polymeric material and said supported catalytic material, and

impinging said gas stream on said substrate surface so as to co-deposit said supported catalytic material particulates and said polymer on said substrate surface.

34. (new) The method of Claim 33 wherein said first region is at an elevated first temperature and said second region is at a second temperature lower than said first region.

35. (new) The method of Claim 34 wherein said catalytic material is produced in said first region by thermal reaction of material that is precursor to said catalytic material.

36. (new) The method of Claim 35 wherein said first temperature is necessary to promote said thermal reaction, said first temperature being inconsistent with stability of said polymeric material, said second temperature being consistent with stability of said polymeric material.

37. (new) The method of Claim 35 wherein said thermal reaction occurs in a flame.

38. (new) The method of Claim 35 wherein quenching means are provided to lower the temperature from said first temperature to said second temperature.

0138A-EC-US

39. (new) The method of Claim 38 wherein said quenching means comprises a quenching fluid introduced into said gas stream downstream of said first region.

40. (new) Apparatus for depositing a catalytic material on a substrate, said apparatus comprising

gas flow means for directing a stream of gas through a first upstream region, a second region downstream of said first region, and a deposition region downstream of said second region,

means for locating a substrate in said deposition region such that the stream of gas impinges on said substrate,

means for introducing particulates of support material into said gas stream at or upstream of said first upstream region,

means at said first upstream region for depositing catalytic material on said support material particulates to produce particulates of supported catalytic material,

at said second region, means for providing a spray of catalytic material so as to mix said polymeric material and said supported catalytic material, and

means to impinge said gas stream on said substrate surface so as to co-deposit said supported catalytic material particulates and said polymer on said substrate surface.

41. (new) Apparatus according to Claim 40 wherein said deposition means at said first location comprises means to introduce a precursor of said catalytic material and means to heat said precursor to produce said catalytic material by thermal reaction.

42. (new) Apparatus according to Claim 40 wherein said heating means is a flame.

43. (new) Apparatus according to Claim 40 including quenching means to lower the temperature from said first region to said second region.

0138A-EC-US

44. (new) Apparatus according to Claim wherein said quenching means comprises means to introduce cooling fluid into said gas stream downstream of said first region.

0138A-EC-US

Appendix B

Replacement Paragraph, page 1, line 24 – page 2, line 8

A prior art method of providing a catalytic layer is to produce a fluid composition, e.g., a solution/suspension, of the layer components, e.g., carbon-supported catalyst dispersed in a solution/suspension of NAFION® (persulfonated polytetrafluoroethylene); apply, e.g., with doctor blades, this fluid to a substrate surface, e.g., the surface of a woven or non-woven carbon cloth; and subsequently dry the fluid to produce the catalytic layer. The surface of the substrate, which comprises a carbon cloth, has an uneven surface. When a layer is formed from a solution and a doctor blade is used in application of the solution, the surface of the catalytic layer away from the substrate is substantially smooth. As the surface of the catalytic layer that contacts the uneven substrate is similarly uneven, the catalytic layer that is formed is of uneven thickness. All portions of the catalytic layer must be sufficiently thick to ensure that substantially all of the gas passing through the layer comes into contact with catalyst. This means that portions of the layer are thicker than is required. As a result, catalytic material, including expensive catalytic metal, is used in excess if applied by this method. Furthermore, thicker portions of a catalytic membrane are less efficient in transporting electrons and in transporting chemicals, such as water, that must be removed from the layer.

0138A-EC-US

Replacement Paragraph, page 2, line 18 – page 2, line 31

U.S. Patent No. 6,403,245 issued 11 June 2002, describes a method of forming catalytic fuel cell layers by co-depositing catalytic particulates, e.g., platinum particulates, formed by combustion chemical vapor deposition (CCVD) and at least a solution containing NAFION (persulfonated polytetrafluoroethylene) and/or suspended carbon particulates. The layers produced by the method of U.S. Patent No. 6,403,245 may be thin and conformal. By “conformal” is meant that the deposited catalytic layers conform in contours to the uneven contours of the substrate on which the layer is deposited, both the surface of the layer on the substrate and the opposite surface of the layer generally conforming in contour to that of the substrate. Such conformal layers exhibit generally uniform thickness that contributes to the efficiency of the layer. While the conformal nature of the layer that forms contributes to the catalytic efficiency of the layer, deposition of layers as described in U.S. Patent No. 6,403,245 has difficulties with high speed, large-scale deposition of catalytic materials and does not account for a variable permeable substrate.

0138A-EC-US

Replacement Paragraph, page 3, line 23 – page 3, line 28

In accordance with another aspect of the invention, carbon-supported catalyst is provided at an upstream location, the carbon-supported catalyst preferably produced *in situ* from carbon particulates and CCVD-produced catalyst. Carrier liquid, such as water and/or alcohol, is substantially removed from the carbon-supported catalyst prior to being introduced into a finely divided spray of a solution of ionomer, such as NAFION® (persulfonated polytetrafluoroethylene). This mixture of ionomer and carbon-supported catalyst is deposited on a substrate surface.

0138A-EC-US

Replacement Paragraph, page 5, line 15 – page 5, line 26

Deposition apparatus described herein is useful for depositing a porous catalytic layer from catalytic material comprising catalytic particulates, e.g., carbon; conductive particulates, e.g., carbon; and polymer, e.g., NAFION® (persulfonated polytetrafluoroethylene). The components of the catalytic material may be supplied and/or generated within the apparatus individually, or two or more of the components may be pre-formed or pre-mixed together. Operation of the apparatus described in reference to Figure 1 is most complex when the individual components of the catalytic material are separately introduced into the apparatus and/or separately formed in the apparatus; accordingly, the apparatus will be first described herein in terms of this mode of catalytic material layer deposition. Subsequently, more simplified operation of the apparatus will be described in reference to depositions in which two or more of the materials are introduced as co-formed and/or co-mixed materials.

0138A-EC-US

Replacement Paragraph, page 6, line 22 – page 7, line 6

In a currently preferred embodiment, nozzles (30), (32), and (34) are used to introduce material into the upstream catalyst-introduction region (22) of the tunnel (21). Through nozzles (30) are introduced a precursor solution for catalyst, such as platinum, ruthenium, gold, palladium, etc. and mixtures of such catalysts. The precursor solution is introduced as a finely divided aerosol is preferably produced by apparatus described in U.S. Patent No. 6,132,656 6,132,653 that is capable of producing sub-micron droplets of solution. The apparatus described in U.S. Patent No. 6,132,656 6,132,653, in which fluid is atomized by passage under pressure through a heated tube, is advantageously utilized in the processes of the invention whenever very fine droplets of fluid are desired. Where very fine droplets are not required, more conventional atomizing apparatus, such as apparatus that atomizes fluids by shear forces, may be used. Thus, then the term “nozzle” is used herein, it is to be understood that nozzle selection will be according to the requirements of the particular deposition method chosen. The various nozzles described herein extend through ports in the housing (19); and the various ports can facilitate alternate types of nozzles according to the requirements of the particular deposition method or can be selectively closed off if not required for the particular deposition method.

0138A-EC-US

Replacement Paragraph, page 7, line 7 – page 7, line 15

Using the apparatus described in U.S. Patent No. ~~6,132,656~~ 6,132,653 in conjunction with dilute solutions of catalyst precursor, very tiny particulates of catalyst may be produced, e.g., having mean particulate diameters in the range of from about 2 nanometers to about 200 nanometers. The carrier liquid in the solution is a flammable liquid or mixture of flammable liquids and the atomized droplets are ignited to produce CCVD flames (40) as described in U.S. Patent No. 5,652,021. Precursor chemicals for Pt, Ru, Au, Cd, and a variety of other metals are described, for example, in U.S. Patent No. 6,208,234. The teachings of all patents and applications discussed herein are incorporated by reference. The material used as the catalyst is not limited and any effective material may be used.

0138A-EC-US

Replacement Paragraph, page 7, line 26 – page 7, line 26

The apparatus in above-discussed U.S. Patent No. ~~6,132,656~~ 6,132,653 atomizes a fluid by passing them through a tubular pressurized region and heating the fluid while in this tubular portion. When the fluid exits the tube, the pressure drops; the fluid rapidly atomizes into very small droplets, and evaporation of liquid components from the tiny droplets happens very rapidly. When such apparatus is used for one or more of the fluids, the vaporizing liquid(s) contribute significantly to the gas through-put of the apparatus, it being appreciated that gas has a volume about 3 orders of magnitude than liquid. By use of an elevated temperature tunnel (21) in which liquid(s) is almost instantaneously turned to gas in the apparatus, helping to maintain separation of spray particles and aiding in drying. This contributes to deposition efficiency of the apparatus. A flux of layer-forming particulates is produced within the tunnel (21).

0138A-EC-US

Replacement Paragraph, page 9, line 29 – page 10, line 13

The electrode substrate is generally porous, because, when used in a fuel cell, gases and liquids are generally required to pass through the substrate. The substrate may, for example, be a woven or non-woven graphite fiber cloth. A currently preferred substrate is a woven graphite fiber cloth that is coated on its deposition surface with a mixture of carbon particulates and ~~PTFE~~ polytetrafluoroethylene (TEFLON®) particulates. The porosity, at least prior to deposition, is sufficiently porous for gases to pass through the substrate. To draw gas-carried, layer-forming material to the substrate surface (12) and into substrate pores, a vacuum source (23) is disposed below the passing substrate (14) in the gas flow path from the tunnel. To allow gas flow through the substrate, the substrate may be supported at its edges by a frame and/or may rest on a screen. The application of vacuum through the substrate (14) results in higher deposition efficiency, i.e., a higher percentage of membrane-forming material depositing on the substrate surface; filling in of pores that are overly large for fuel cell operation, and better adhesion of the deposited layer to the substrate. The intensity of the vacuum is adjusted according to a variety of factors, including gas flow rate above the substrate, pore size and porosity of the substrate, and composition of the layer-forming material being deposited on the substrate surface (12).

0138A-EC-US

Replacement Paragraph, Page 18, lines 20-25

The most common proton-transfer media for use in proton-conducting membranes are ionomers, such as a persulfonated perfluoro-ether polytetrafluoroethylene, c.g., that sold as Nafion®. Such ionomers can be deposited from a solution that is co-deposited with CCVD flame-produced material. However, proton-conducting material may be an inorganic material, such as zirconium phosphate or silicotungstic acid, that can be deposited directly from a CCVD flame along with the water-binding material.

0138A-EC-US

Replacement Paragraph, page 18, line 26 – page 19, line 8

The water-binding material that provides for water retention within the proton-conducting membrane is generally a water-binding oxide or mixed oxide, such as silica or a zeolite. Zeolites are naturally hydrated silicates of aluminum plus sodium and/or calcium and/or potassium. Such water-binding oxides are easily produced by CCVD flames from a precursor solution containing chemical precursors of the mixed oxides. When co-deposited with a proton-transfer material, either an ionomer, such as Nafion® (persulfonated polytetrafluoroethylene), from a co-deposition solution, or an inorganic material co-deposited from a CCVD flame, the water-binding oxides are deposited in very small domains that are homogeneously and intimately admixed with the proton-transfer material and serve to maintain adequate hydrolysis levels of the membrane. The weight ratio of the water-binding material to the proton-transfer material in the membrane may vary over a wide ratio, depending upon the particular materials selected and the desired operating temperature of the fuel cell. Typically the weight ratio of the water-binding material to the proton-transfer material ranges from about 1:20 to about 2:1.

0138A-EC-US

Replacement Paragraph, page 19, lines 16-19

Most commonly, however, the proton-transfer material for the proton-conducting membrane is an ionomer, Nafion® (persulfonated polytetrafluoroethylene) being the currently preferred ionomer. In such case, the ionomer is deposited from a non-flame spray(s) of ionomer solution, while the inorganic water-binding material is deposited from a CCVD flame.

0138A-EC-US

Replacement Paragraph, page 21, lines 5-16

While the catalyst, particularly platinum, may be distributed throughout the membrane, it is preferred that it be deposited as a layer 113 intermediate layers 115, 117 of proton-transfer material, such as Nafion® (persulfonated polytetrafluoroethylene) as shown in Fig. 6. Also, preferably, this layer is deposited closer to the cathode (oxygen) side than to the anode (hydrogen) side. This is because hydrogen, being a much smaller molecule than oxygen, has a greater conduction rate through the membrane. The location of the thin layer of catalyst can be placed at an optimal location within the membrane to account for the higher relative migration of hydrogen relative to oxygen. It is also within the scope of the invention to deposit catalytic material throughout the proton-transfer material and also provide a discrete film or layer of the catalytic material. The film or layer 113 of catalytic material is typically very thin, e.g., between about 10 and about 80 nanometers. It must be sufficiently porous to provide for proton-transfer therethrough.

0138A-EC-US

Replacement Paragraph, page 22, lines 3-21

When an intermediate layer 113 or 119 is formed for proton-conducting membranes 82' or 82", respectively, various orders of fabrication may be used. The membrane 113 or 119 may be deposited directly on a layer of proton-transfer material and a second layer of proton-transfer material subsequently deposited thereon. Such alternate deposition may be effected with the apparatus of Fig. 7 by alternately using the non-flame spray flame to deposit ionomer and using the flame, in conjunction with a re-direct spray, e.g., water, from the non-flame spray, to deposit the material of the intermediate layer 113 or 119. Alternatively, the intermediate layer 113 or 119 may be deposited on a material, such as a metal foil or a polytetrafluoroethylene (Teflon®) substrate and subsequently transferred to an ionomer layer (e.g., Nafion® (persulfonated polytetrafluoroethylene)) in a "decals transfer" technique. A subsequent layer of proton-transfer material would then be deposited on top of this layer. Optimally, the intermediate layer 113 or 119 are deposited with the methods and apparatus described with respect to Figs. 1-3. In this case, after the ECL is deposited, the proton-exchange membrane (PEM) material is deposited to fill all existing permeability of the layer. In such a system, the PEM (56) may be thinner or omitted and the two sides combined directly. The ECLs can be formed from one Tunnel (21) and the intermediate layer from another tunnel (21) in the correct sequence. Some amount of PEM can be formed between the ECL and the intermediate layer via another tunnel connection.